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14. ABSTRACT

This grant supported two postdoctoral fellows who worked on a variety of problems related to the prediction of materials properties from first principles. Projects included optimization of a parallel code for calculating magnetic anisotropy parameters; application of the method to molecule-based magnets; studies of structural and electronic properties of interfaces between ferromagnets and semiconductors; and investigation and quantification of vibrational contributions to the Van der Waals interaction between molecules.

15. SUBJECT TERMS

Density functional theory; magnetic anisotropy; molecular magnets; magnetic junctions

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Prediction of Magnetic and Electronic Phenomena in Molecular-Assembled Crystals

Final Technical Report

Summary

This grant has supported the salary and research expenses of two post-doctoral fellows, Tunna Baruah (14 months) and Kyungwha Park (14 months), and summer salary for the PI.

The density-functional code NRLMOL was reprogrammed to allow efficient calculation of magnetic anisotropy energies and principal axes of magnetization on large-scale parallel platforms. The code has been applied to study the properties of a variety of molecule-based magnets. In addition, a study of the electronic structure and Schottky barrier formation at interfaces between magnetic materials and semiconductors was performed. Further, the vibrational contribution to intermolecular Van der Waals interaction was formulated and investigated using density-functional theory calculations.

Technical Projects

Parallel Computation of Magnetic Anisotropy

A method for the calculation of the second-order magnetic anisotropy energies and principal axes of magnetization was optimized for parallel platforms. This involved addition of more than 1500 lines of code into NRLMOL, a local-orbital-based density functional package. Parallelization has been done with standard public-domain message-passing-interface protocols, and the program currently runs on Linux-based beowulf clusters, IBM SP's and silicon graphics SGIs.

Properties of Nanomagnets

Density functional theory has been used to study the structural, electronic, magnetic, and vibrational properties of a variety of molecule-based magnets, including Mn_{10} and transition-metal dicyanamides. Density functional theory with the exchange and correlation interaction treated at the level of the generalized gradient approximation was found to be very accurate for the determination of geometries, vibrational energetics, and magnetic order. For magnetic anisotropy energies, calculations agreed reasonably well with available experimental data. For the single-molecule magnet Mn_{12}, it was shown that for optimization of anisotropy energies, molecular structure and molecular environment are important considerations.

Magnetic Junctions

The electronic and magnetic properties of Fe/GaAs(001) magnetic junctions were investigated using first-principles density-functional calculations. Abrupt and intermixed interfaces were considered, and the dependence of charge transfer, magnetization profiles, Schottky barrier heights, and spin polarization of densities of states on interface structure was studied. With As-termination, an abrupt interface with Fe is favored, while Ga-terminated GaAs favors the formation of an intermixed layer with Fe. The Schottky barrier heights are particularly sensitive to the abruptness of the interface. A significant density of states in the semiconducting gap arises from metal interface states. These spin-dependent interface states lead to a significant minority spin polarization of the density of states at the Fermi level that persists well into the semiconductor, providing a channel for the tunneling of minority spins through the Schottky barrier.

Vibrational Van der Waals Interaction

The van der Waals interaction can be caused by either ionic vibrations or instantaneous electronic motion relative to the atomic center. In this study, the vibrational contribution to the van der Waals interaction was formulated by considering the interaction between induced dipoles caused by the infrared-active normal modes of a neutral molecule. Using the derived formula, the contribution was quantified, within the density-functional theory formalism, using a screened vibrational polarizability. Application to several neutral nonpolar dimers indicated vibrational contributions substantially smaller than electronic contributions. It was shown that the ratio of the vibrational to electronic contributions depends strongly on the ratio of the screened vibrational to electronic polarizabilities and on the ratio of the frequency of the strongest infrared-active mode to an ionization energy.

Publications

J. Kortus, M. R. Pederson, **T. Baruah**, *et al.*, "Density functional studies of single molecule magnets," Polyhedron **22**, 1871 (2003).

T. Baruah and M. R. Pederson, "Density functional study of the conformers of Co_4-based single-molecule magnet," Int. J. Quantum Chem. **93**, 324 (2003).

K. Park, **T. Baruah**,*et al.* "Second-order transverse magnetic anisotropy induced by disorder in the single-molecule magnet Mn_{12}," Phys. Rev. B **69**, 144426 (2004)

K. Park and M. R. Pederson, "Effect of extra electrons on the exchange and magnetic anisotropy in the anionic single-molecule magnet Mn_{12}," Phys. Rev. B **70**, 054414 (2004)

D. O. Demchenko, **A. Y. Liu**, *et al.* "Electronic structure of the transition-metal dicyanamides $M[N(CN)_2]_2$ (M=Mn,Fe,Co,Ni,Cu)," Phys. Rev. B **69**, 205105 (2004).

K. Park, **A. Y. Liu**, and M. R. Pederson, "Comparison of vibrational and electronic contributions to Van der Waals interactions," Phys. Rev. B **73**, 205116 (2006).

D. O. Demchenko and **A. Y. Liu**, "Influence of interface structure on electronic properties and Schottky barriers in Fe/GaAs magnetic junctions," Phys. Rev. B **73**, 115332 (2006).

I. Schnell, I. I. Mazin, and **A. Y. Liu**, "Unconventional superconducting pairing symmetry induced by phonons," Phys. Rev. B **74**, 184503 (2006).

K. Park *et al.*, "Electronic structure and vibrational spectra of C_2B_{10}-based clusters and films," Phys. Rev. B **74**, 035109 (2006).